

Deterministic single-atom excitation via adiabatic passage and Rydberg blockade

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We propose to use adiabatic rapid passage with a chirped laser pulse in the strong dipole blockade regime to deterministically excite only one Rydberg atom from randomly loaded optical dipole traps or optical lattices. The chirped laser excitation is shown to be insensitive to the random number N of the atoms in the traps. Our method overcomes the problem of the \sqrt{N} dependence of the collective Rabi frequency, which was the main obstacle for deterministic single-atom excitation in the ensembles with unknown N , and can be applied for single-atom loading of dipole traps and optical lattices.

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I. INTRODUCTION

Dipole blockade of the laser excitation of neutral atoms to highly excited Rydberg states [1] opens up new opportunities for entanglement engineering and quantum information processing [2]. In mesoscopic ensembles of strongly interacting neutral atoms the dipole blockade manifests itself as a suppression of the excitation of more than one atom by narrow-band laser radiation, due to the shifts of the Rydberg energy levels induced by long-range interactions [1]. Recently, dipole blockade was observed for two Rb atoms trapped in two spatially separated optical dipole traps [3–5].

The deterministic excitation of a single atom from a mesoscopic ensemble into a Rydberg state using dipole blockade can be exploited in a variety of applications, as the creation of quantum information processor for photons [6] or single-atom loading of optical lattices or optical dipole traps [7], which is of crucial importance for the development of quantum registers [2], single-photon sources [8], high-precision metrology in optical lattice clocks [9], and phase transitions in artificial solid structures with Rydberg excitations [10].

Single-atom loading remains a challenge since no simple and reliable method for the preparation of the optical lattice with single occupancy at each site is available yet. So far, only the Mott insulator regime in Bose-Einstein condensates (BEC) has demonstrated its ability to provide single-atom loading of large-scale optical lattices [11]. However, obtaining a BEC is a complicated and slow procedure, which may be not well suited for fast quantum computation. Another approach for single-atom loading of multiple sites is the exploitation of a collisional blockade mechanism [12], but it suffers from the low loading efficiency for large arrays. Recently, the fi-

delity of single atom loading of 82.7% was demonstrated using light-assisted collisions [13]. Although being an important step forward, this fidelity is not enough for the creation of a scalable quantum register [2].

Highly excited Rydberg atoms with the principal quantum number $n \gg 1$ [14] can be used to implement fast quantum logic gates [1, 15]. These atoms exhibit strong dipole-dipole interaction at distances that can be as large as a few microns. Therefore, dipole-dipole interaction can also be used in schemes for single-atom loading of optical lattices and traps arrays, since the interatomic spacing in lattices sites lies in the micron range.

The first proposal for single-atom loading exploiting a dipole blockade of the laser excitation of mesoscopic ensembles of N cold ground-state atoms [1, 16] was formulated in Ref. [7]. A strong dipole blockade was suggested to provide deterministically a single Rydberg atom, while the remaining $N - 1$ ground-state atoms could be selectively removed from the lattice site by an additional laser pulse. In Ref. [2] it has been pointed out, however, that this method demands identical initial numbers of cold atoms in each lattice site, because collective Rabi frequency of single-atom excitation depends on N :

$$\Omega_N = \Omega_1 \sqrt{N} \quad (1)$$

(here Ω_1 is the Rabi frequency for a single atom). This requirement is not fulfilled in optical lattices, which are loaded from cold atom clouds at random and typically have a Poissonian distribution of the number of atoms in each site [17].

Full dipole blockade ensures that an ensemble of N atoms shares a collective single excitation oscillating between the ground and Rydberg state at the Rabi frequency Ω_N [18]. For a given number N of atoms experiencing full dipole blockade, it is possible to excite a single atom into the Rydberg state using a monochromatic laser

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π -pulse of duration

$$\tau_N = \pi / (\Omega_1 \sqrt{N}). \quad (2)$$

However, in randomly loaded traps or optical lattices the atom-number uncertainty for the Poissonian statistics is $\Delta N \approx \sqrt{N}$, where N is the mean number of atoms in the traps. Therefore, although the suppression of the excitation of more than one atom in the trap is guaranteed in the full dipole blockade regime, the value of τ_N required for the deterministic single-atom excitation is uncertain due to the uncertainty of N in the individual traps [2].

Adiabatic passage by sweeping the laser frequency through the resonance [19] or by counter-intuitive sequence of two monochromatic pulses known as Stimulated Raman Adiabatic Passage or STIRAP [20] is widely used to obtain a population inversion in multi-level systems due to their insensitivity to the Rabi frequencies of the particular transitions, provided the adiabaticity condition is satisfied [20, 21]. We therefore may expect that adiabatic passage is also suitable to the blocked ensembles with unknown number of atoms. However, recently it has been found that STIRAP with zero detuning from the intermediate state does not provide deterministic single-atom excitation in a blocked ensemble [22]. In this work we propose to deterministically excite a single Rydberg atom using a chirped laser pulse or off-resonant STIRAP.

II. DETERMINISTIC SINGLE-ATOM EXCITATION WITH CHIRPED LASER PULSES

The time dependence of the amplitude and frequency for the chirped laser pulse is shown in Fig. 1(a). The laser frequency is linearly swept across the resonance during the pulse. In contrast to the results of Ref. [22], we have found that chirped laser excitation deterministically transfers the population of the blocked ensemble into the collective state, which shares a single Rydberg excitation, as shown in Fig. 1(b).

The principle of chirped excitation can be understood as follows [23]. The dressed state energies for a two-level atom are shown in Figs. 1(c)-1(e) for laser detunings $\Delta < 0$, $\Delta \sim 0$ and $\Delta > 0$, respectively. At large negative detuning $\Delta < 0$ the energy of the unperturbed ground state g is close to the energy of the dressed state g_- , whereas at large positive detuning $\Delta > 0$ the energy of the dressed state r_- lies close to the energy of the unperturbed Rydberg state r . Initially, only the ground g_- state is populated [Fig. 1(c)]. When the laser frequency is swept across the resonance [Fig. 1(d)] the system adiabatically follows the dressed $\{-\}$ state of the dressed system, and therefore populates the Rydberg state r_- after the end of the chirped laser pulse [Fig. 1(e)].

In order to show that chirped laser excitation in the blocked ensemble is insensitive to N , we have performed numerical simulations of the dipole blockade for

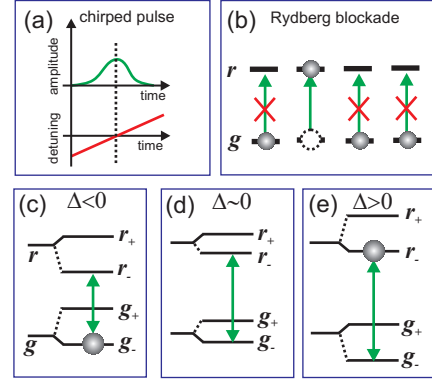


FIG. 1: (Color online). (a) The time dependence of the amplitude and detuning from the resonance with atomic transition for a chirped pulse; (b) Scheme of the deterministic single-atom excitation from ground state g to Rydberg state r in a blocked Rydberg ensemble. (c),(d),(e) Scheme of the adiabatic rapid passage. Energies of the dressed states are shown for laser detunings (c) $\Delta < 0$, (d) $\Delta \sim 0$, and (e) $\Delta > 0$. Laser frequency is rapidly chirped across the resonance. The population is transferred from state g_- to state r_- independently of the Rabi frequency.

$37P_{3/2}$ Rydberg state in Rb atoms. Rydberg atoms in an identical state nL may interact via a Förster resonance if this state lies midway between two other levels of the opposite parity [24]. The $37P_{3/2}$ state has a convenient Stark-tuned Förster resonance $37P_{3/2} + 37P_{3/2} \rightarrow 37S_{1/2} + 38S_{1/2}$, which we studied earlier in detail [25, 26].

We consider an excitation of the $37P_{3/2}$ state by a linearly-chirped Gaussian laser pulse (Fig. 2). In the time domain its electric field is expressed as

$$E(t) = E_0 \exp\left[\frac{-t^2}{2\tau^2}\right] \cos\left[\omega_0 t + \alpha \frac{t^2}{2}\right]. \quad (3)$$

Here E_0 is the peak electric field at $t = 0$, ω_0 is the frequency of the atomic transition, $\tau = 1 \mu\text{s}$ is the half-width at $1/e$ intensity [Fig. 2(a)], and α is the chirp rate [21]. We choose E_0 to be such as to provide a single-atom peak Rabi frequency $\Omega_1/(2\pi) = 2 \text{ MHz}$ or $\Omega_1/(2\pi) = 0.5 \text{ MHz}$ at the $5S \rightarrow 37P_{3/2}$ optical transition in Rb atoms. For convenience, the central frequency of the laser pulse is taken to be exactly resonant with the atomic transition at the maximum of the pulse amplitude. The atoms begin to interact with the laser radiation at $t = -4 \mu\text{s}$.

The adiabaticity condition for a chirped pulse exciting a single Rydberg atom is given by [21]:

$$|d\Delta/dt| \ll \Omega_1^2 \quad (4)$$

For $N > 1$ the collective Rabi frequency $\Omega_N = \Omega_1 \sqrt{N}$ grows with N . Hence, we must only fulfill the adiabaticity condition for the excitation of a single atom.

The envelope of the laser pulse is a Gaussian that ensures the adiabatic switching of the laser-atom interaction. Figure 2(b) shows the calculated spectra of

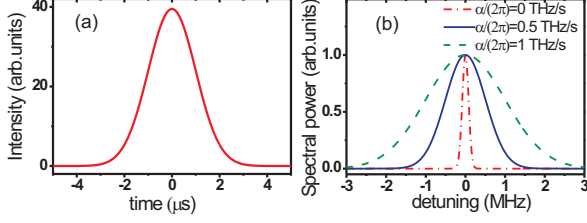


FIG. 2: (Color online). (a) Envelope and (b) spectrum of the chirped Gaussian laser pulses with the chirp rates $\alpha/(2\pi) = 0.5$ THz/s (solid line) and $\alpha/(2\pi) = 1$ THz/s (dashed line) used in numerical calculations. The spectrum of the unchirped laser pulse of the same duration is shown for reference (dash-dotted line, height rescaled for clarity).

the laser pulses with $\alpha/(2\pi) = 0.5$ THz/s (solid line) and $\alpha/(2\pi) = 1$ THz/s (dashed line). The spectrum is broadened to the FWHM of $\Delta\omega/(2\pi) = 1.2$ MHz at $\alpha/(2\pi) = 0.5$ THz/s and $\Delta\omega/(2\pi) = 2.4$ MHz at $\alpha/(2\pi) = 1$ THz/s due to the frequency chirp, as can be seen from the comparison with the unchirped Gaussian pulse [$\Delta\omega/(2\pi) = 0.2$ MHz] shown as the dash-dotted line in Fig. 2(b). This broadening could affect the dipole blockade efficiency and lead to leakage of the population to the collective states with more than one excitation. We therefore have performed a numerical calculation of the blockade efficiency for chirped laser excitation of an ensemble consisting of $N=1-7$ atoms. The time-dependent Schrödinger equation was solved for the amplitudes of the collective states, taking into account all possible binary interactions between Rydberg atoms [25, 26].

The calculations have been done for the exact Stark-tuned Förster resonance $37P_{3/2} + 37P_{3/2} \rightarrow 37S_{1/2} + 38S_{1/2}$ with zero energy defect which is supposed to be tuned by the electric field of 1.79 V/cm [25]. The numerically calculated time dependencies of the probability P_1 to excite a single Rydberg atom by the chirped laser pulse are shown in Fig. 3 for $\alpha/(2\pi) = 1$ THz/s, $\Omega_1/(2\pi) = 2$ MHz (the left-hand panels) and for $\alpha/(2\pi) = 0.5$ THz/s, $\Omega_1/(2\pi) = 0.5$ MHz (the right-hand panels). Figures 3(a)-3(b) correspond to $N=1$ (a single non-interacting atom) and serve as the references to compare with the interacting atoms. The transition probability in Fig. 3(a) is nearly unity with accuracy better than 0.02%, while in Fig. 3(b) it reaches 0.993 at the end of the laser pulse. However, below we will show that for single-atom excitation at $N > 1$ the conditions of Fig. 3(b) are preferred.

The calculated time dependencies of P_1 in the full dipole blockade regime are shown in Fig. 3 for 2 atoms [(c)-(d)], 5 atoms [(e)-(f)], and 7 atoms [(g)-(h)]. The N atoms were randomly located in a $L \times L \times L$ μm^3 cubic volume with $L = 1$ μm . The full blockade regime was evidenced by complete suppression of the probability to excite more than one atom. The calculations have shown that the fidelity P_1 of the population inversion at $t = 4$ μs reaches 99% regardless of N . This is the main

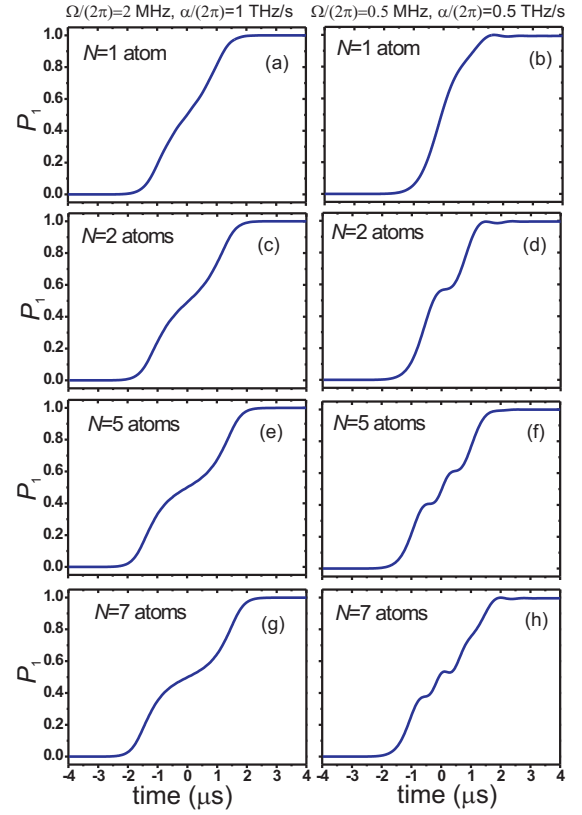


FIG. 3: (Color online). Time dependence of the probability P_1 to excite a single Rb($37P_{3/2}$) atom in a trap containing (a)-(b) 1 atom, (c)-(d) 2 atoms, (e)-(f) 5 atoms, and (g)-(h) 7 atoms by chirped Gaussian laser pulses. In the left-hand panels the chirp rate is $\alpha/(2\pi) = 1$ THz/s and Rabi frequency is $\Omega_1/(2\pi) = 2$ MHz. In the right-hand panels the chirp rate is $\alpha/(2\pi) = 0.5$ THz/s and Rabi frequency $\Omega_1/(2\pi) = 0.5$ MHz. The calculations have been done for the exact Stark-tuned Förster resonance $37P_{3/2} + 37P_{3/2} \rightarrow 37S_{1/2} + 38S_{1/2}$ with zero energy defect. The atoms are randomly placed in the cubic interaction volume of the size $L = 1$ μm .

result of this article that confirms that our proposal can be implemented in practice.

Surprisingly, the effect of chirped laser excitation in the blockaded ensemble is completely different from the effect of on-resonant STIRAP, discussed in Ref. [22]. The scheme of the three-level ladder system interacting with the two laser fields with Rabi frequencies Ω_s and Ω_c is shown in Fig. 4(a). We consider an exact two-photon resonance for the ground state g and Rydberg state r , while the intermediate excited state e has in general a variable detuning δ from the resonance both with Ω_s and Ω_c fields (but with the opposite signs). The time sequence of the two Gaussian pulses with halfwidths τ_c and τ_s at $1/e$ intensity and time interval ΔT is shown in Fig. 4(b). For simplicity, we have chosen identical Rabi frequencies $\Omega_s/(2\pi) = \Omega_c/(2\pi) = 10$ MHz. The pulse halfwidths $\tau_c = \tau_s = 1$ μs were also identical to the half of the interval between the pulses $\Delta T = 2$ μs [20].

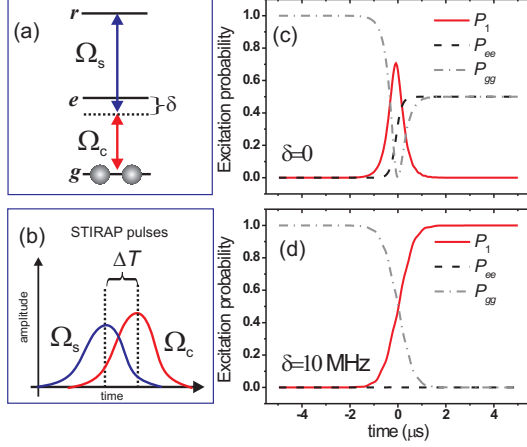


FIG. 4: (Color online). (a) Scheme of the three-level ladder system interacting with the two laser fields with Rabi frequencies Ω_s and Ω_c and detuning from the intermediate state δ . (b) Time sequence of pulses for STIRAP. The delay between the two pulses is ΔT . (c) Time dependencies of the probabilities P_{gg} , P_{ee} and the probability to excite a single Rydberg atom P_1 for on-resonant STIRAP with $\delta = 0$. (d) Time dependencies of the probabilities P_{gg} , P_{ee} and the probability to excite a single Rydberg atom P_1 for off-resonant STIRAP with $\delta = 10$ MHz.

We have calculated the excitation probabilities of the collective states of an ensemble consisting of two interacting atoms in a full blockade regime. The time dependencies of the probability of single-atom Rydberg excitation P_1 (solid line) and the probabilities P_{ee} (both atoms in the lower excited state, dashed line), P_{gg} (both atoms in the ground state, dash-dotted line) are shown for on-resonant STIRAP with $\delta = 0$ in Fig. 4(c) and for off-resonant STIRAP with detuning from the intermediate state $\delta = 10$ MHz in Fig. 4(d). Figure 4(c) qualitatively reproduces the results of Ref. [22] for two atoms. After the end of the adiabatic passage the population is redistributed between g and e states, while Rydberg state r remains unpopulated. We have found that the observed breakdown of STIRAP in the blockaded ensemble results from the destructive interference of laser-induced transitions in the quasimolecule consisting of interacting atoms, and it can be avoided by an increase of the detuning from the intermediate state, which finally makes STIRAP equivalent to chirped excitation [27]. Figure 4(d) shows that as well as chirped excitation, off-resonant STIRAP can be used for deterministic excitation of a single Rydberg atom.

III. FIDELITY OF DETERMINISTIC SINGLE-ATOM EXCITATION

The main limitation of the proposed method is a possible breakdown of the full dipole blockade in the realis-

tic experimental conditions. The N atoms in an optical dipole trap have a finite temperature and are located randomly due to atomic motion. The blockade breakdown for two interacting Rydberg atoms can be caused by the weakness of dipole-dipole interaction between remote atoms or by more complicated mechanisms, including zeros of Förster resonances [28] and destructive interference in many-atom ensembles [29].

The fidelity of the single-atom excitation can be defined as the probability P_1 to have exactly one atom excited at the end of the laser pulse. We have numerically calculated P_1 for various sizes of the atomic sample in an optical dipole trap for chirped laser excitation. The N atoms were randomly located in a $L \times L \times L$ μm^3 cubic volume. The value of P_1 was averaged over $\sim 10^4$ random spatial configurations.

The dependencies of P_1 on L at the end of the laser pulse ($t = 4$ μs) are shown in Fig. 5(a) for $\alpha/(2\pi) = 1$ THz/s, $\Omega_1/(2\pi) = 2$ MHz and 2 (squares), 3 (circles), 4 (triangles), and 5 (rhombs) atoms. For $L > 1$ μm we have found that P_1 reduces as L increases, mostly due to the fluctuations of the spatial positions of the atoms in a disordered sample [26]. More surprisingly, we have found that P_1 depends on N in a counter-intuitive way: it drops as N increases. This observation is presumably due to the quantum interference between different energy exchange channels in many-atom ensembles, which was discussed in Ref. [29]. Our Monte Carlo numerical simulations in Fig. 5(a) and (b), which accounted also for the Förster zeros [28], have shown that they do not affect the dipole blockade even for atoms randomly placed within the interaction volume, if the Rydberg interactions are strong enough, i.e., when the blockade shift is larger than the laser linewidth.

The same calculations have been done for $\alpha/(2\pi) = 0.5$ THz/s and $\Omega_1/(2\pi) = 0.5$ MHz [Fig. 5(b)]. Although in these conditions P_1 also reduces with the increase of L and N , the efficiency of the dipole blockade is better, than in Fig. 5(a). For all N we have found that in Fig. 5(b) the fidelity $P_1 \geq 0.95$ is achieved at $L \leq 2$ μm , while in Fig. 5(a) a localization $L \leq 1$ μm is required. This difference can be crucial in the real experimental conditions, since it is difficult to localize the atoms in the volumes of the size comparable with optical wavelengths.

The increase of the chirp rate and Rabi frequency may be desirable to reduce the excitation time and to avoid the errors due to finite lifetimes of Rydberg states [30]. We have found, however, that such an increase would decrease the blockade efficiency, which depends on both Ω_1 and α . To find the optimal values of Ω_1 and α for the two frozen atoms in the full blockade regime, which can be modeled simply by increasing the effective Rabi frequency $\Omega_2 = \Omega_1\sqrt{2}$ in a single two-level atom. This dependence is presented in Fig. 5(c) as a density plot, as in Ref. [21]. The light areas in Fig. 5(c) show the regions where $P_1 \approx 1$. The periodic structure across the $\alpha = 0$ axis represents the coherent Rabi oscillations

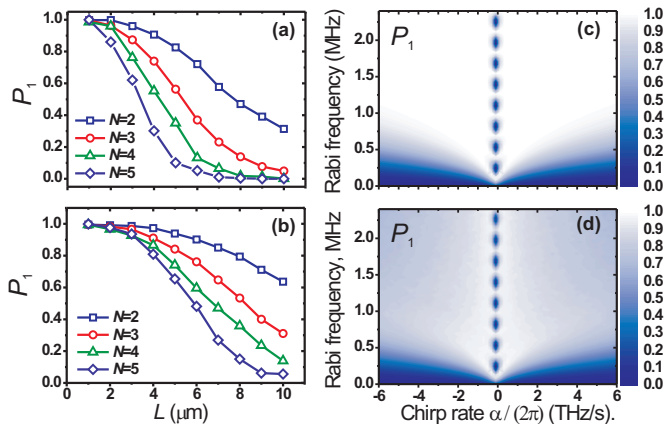


FIG. 5: (Color online). (a) Dependencies of the probability P_1 to excite a single $\text{Rb}(37P_{3/2})$ atom on the size L of the cubic interaction volume for 2 (squares), 3 (circles), 4 (triangles), and 5 (rhombs) randomly positioned atoms at the chirp rate $\alpha/(2\pi) = 1$ THz/s and Rabi frequency $\Omega_1/(2\pi) = 2$ MHz. (b) The same dependencies at the chirp rate $\alpha/(2\pi) = 0.5$ THz/s and Rabi frequency $\Omega_1/(2\pi) = 0.5$ MHz. (c) Dependence of P_1 on Rabi frequency and chirp rate for the two frozen atoms in the full blockade regime. (d) Monte-Carlo simulation of the same dependence for the two interacting atoms, randomly placed in the cubic volume with $L = 4 \mu\text{m}$. The calculations have been done for the exact Stark-tuned Förster resonance $37P_{3/2} + 37P_{3/2} \rightarrow 37S_{1/2} + 38S_{1/2}$ with zero energy defect.

at the $n\pi$ laser pulses with zero chirp. The area of the robust rapid adiabatic passage at $\alpha \ll 1/\tau^2$ is limited by the adiabaticity condition of Eq. (4). However, a possible breakdown of the dipole blockade adds more restrictions on the values of Ω_1 and α . The dependence of P_1 on Ω_1 and α for the two interacting atoms randomly placed in the cubic volume with $L = 4 \mu\text{m}$ is shown in Fig. 5(d). The probability P_1 drops with the increase of both Ω_1 and α . However, it remains nearly constant in the region between the coherent and adiabatic regimes with small chirp rate $0.3 \text{ THz/s} \leq |\alpha/(2\pi)| \leq 0.7 \text{ THz/s}$. For our Förster resonance $37P_{3/2} + 37P_{3/2} \rightarrow 37S_{1/2} + 38S_{1/2}$ and the pulse width $\tau = 1 \mu\text{s}$ we have also found the optimal Rabi frequency to be $0.4 \text{ MHz} \leq \Omega_1/(2\pi) \leq 0.6 \text{ MHz}$.

IV. DISCUSSION

We now briefly discuss a possible experimental implementation of the method. Micrometer-sized dipole

traps can be used to store several atoms and to control their positions. By tuning the loading parameters, one can achieve control on the average number of the loaded atoms, which can be limited to $\bar{N} \approx 1 - 10$. The typical lifetimes of these traps can easily reach hundreds of microseconds, and an effective atomic confinement of few hundred nanometers can be achieved in all dimensions [31]. Therefore a high fidelity of the single-atom excitation should be expected in microscopic dipole traps loaded with small number of atoms.

The intense laser field of the dipole trap induces position-dependent light shifts of the atomic energy levels and can also photoionize Rydberg atoms. The effect of light shifts can be suppressed by using the trapping light with a magic wavelength, which matches light shifts of the ground and Rydberg states [32]. The photoionization can substantially reduce effective lifetimes of Rydberg atoms [33] and this effect cannot be suppressed with a magic wavelength. We therefore suggest to avoid both light shifts and photoionization by temporarily switching the dipole trap off, provided the atom temperatures are sufficiently low ($< 50 \mu\text{K}$) to make the subsequent recapture possible.

To conclude, we have proposed a method for the deterministic single-atom excitation to a Rydberg state in mesoscopic ensembles of interacting atoms. Chirped laser excitation and off-resonant STIRAP in the full blockade regime have been shown to be insensitive to the number of interacting atoms. This method is well suited to prepare a collective excited state in a small dipole-blockaded sample of atoms loaded in micrometer sized dipole traps with high fidelity. It could further be used for selective single-atom loading of optical lattices and dipole trap arrays, which are initially loaded with an unknown number of atoms.

Our method opens the way to the implementation of scalable quantum registers and single-photon sources for quantum information processing with neutral atoms.

V. ACKNOWLEDGEMENTS

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